### FATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

## Commissioner NOTIFICATION OF ELECTION US Department of Commerce United States Patent and Trademark (PCT Rule 61.2) Office, PCT 2011 South Clark Place Room CP2/5C24 Arlington, VA 22202 ETATS-UNIS D'AMERIQUE Date of mailing (day/month/year) in its capacity as elected Office 30 May 2001 (30.05.01) International application No. Applicant's or agent's file reference PCT/CA00/01017 755-208/DPC International filing date (day/month/year) Priority date (day/month/year) 01 September 2000 (01.09.00) 03 September 1999 (03.09.99) Applicant HOJABR, Sassan et al 1. The designated Office is hereby notified of its election made: X in the demand filed with the International Preliminary Examining Authority on: 09 March 2001 (09.03.01) in a notice effecting later election filed with the International Bureau on: 2. The election was not made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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# **PCT**

### INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicants or agents tile reference	(Form PCT/ISA	n of Transmittal of International Search Report V220) as well as, where applicable, item 5 below.
755-208/DPC	ACTION	
International application No.	International filing date (day/month/year)	(Earliest) Priority Date (day/month/year)
PCT/CA 00/01017	01/09/2000	03/09/1999
Applicant		
E. I. DU PONT DE NEMOURS	AND COMPANY et al.	
This International Search Report has bee according to Article 18. A copy is being tra		uthority and is transmitted to the applicant
	a copy of each prior art document cited in the	nis report.
Basis of the report		
With regard to the language, the language in which it was filed, unit	international search was carried out on the t ess otherwise indicated under this item.	easis of the international application in the
the international search w Authority (Rule 23.1(b)).	as carried out on the basis of a translation o	f the International application furnished to this
<ul> <li>With regard to any nucleotide an was carried out on the basis of the</li> </ul>	d/or amino acid sequence disclosed in the	international application, the international search
	nal application in written form.	
filed together with the inte	rnational application in computer readable for	orm.
furnished subsequently to	this Authority in written form.	
furnished subsequently to	this Authority in computer readble form.	
	sequently furnished written sequence listing	does not go beyond the disclosure in the
I — · · ·		n is identical to the written sequence listing has been
2. Certain claims were fou	nd unsearchable (See Box I).	
3. Unity of invention is lac	king (see Box II).	
With regard to the title,		
X the text is approved as su	bmitted by the applicant.	
the text has been establis	hed by this Authority to read as follows:	
<ol><li>With regard to the abstract,</li></ol>		
X the text is approved as su		
the text has been establis within one month from the	hed, according to Rule 38.2(b), by this Author date of mailing of this International search r	ority as it appears in Box III. The applicant may, eport, submit comments to this Authority.
6. The figure of the drawings to be publ	ished with the abstract is Figure No.	1
X as suggested by the appli	cant.	None of the figures.
because the applicant fail	ed to suggest a figure.	_
because this figure better	characterizes the invention.	

### PATENT COOPERATION TREATY

**PCT** 

REC'D 12 DEC 2001
WIPO PCT

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

App	licant's	or age	ent's file reference	FOR FURTUER ACTION		ation of Transmittal of International
755	5-208/	OPC		FOR FURTHER ACTION	Preliminary	Examination Report (Form PCT/IPEA/416)
Inte	matlona	appl	ication No.	International filing date (day/month	/year)	Priority date (day/month/year)
PC	T/CA0	0/01	017	01/09/2000		03/09/1999
	mationa 9J123/		ent Classification (IPC) or nat	ional classification and IPC		
App	licant					
E. I	. DU F	ON	T DE NEMOURS AND	COMPANY et al.		
2.	This F	trans EPC nis resen a ee R	smitted to the applicant a PRT consists of a total of sport is also accompanied imended and are the bas	ccording to Article 36.  7 sheets, including this cover st t by ANNEXES, i.e. sheets of thi is for this report and/or sheets of the Administrative Instruction	neet. e description ontaining re	mational Preliminary Examining Authority  n, claims and/or drawings which have clifications made before this Authority e PCT).
3.	This re	eport	Basis of the report Priority Non-establishment of o Lack of unity of invention		•	and industrial applicability
	V	23		ns suporting such statement	ioveity, inve	move step of moustrial applicability;

Date of submission of the demand	Date of completion of this report
09/03/2001	11.12.2001
Name and mailing address of the international	Authorized officer
preliminary examining authority:  European Patent Office - P.B. 5818 Patentlaan 2  NL-2280 HV Rijswijk - Pays Bas  Tel. +31 70 340 - 2040 Tx: 31 651 epo nl	Schmidt, H
Fax: +31 70 340 - 3016	Telephone No. +31 70 340 2461

☐ Certain documents cited

☐ Certain defects in the international application ☐ Certain observations on the international application

### INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/CA00/01017

### I. Basis of the report

and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)): Description, pages: 1-17 as originally filed Claims, No.: 2-8.10-12.14-25 as originally filed 1.9.13 as amended under Article 19 2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item. These elements were available or furnished to this Authority in the following language: . . which is: the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)). the language of publication of the international application (under Rule 48.3(b)). the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3). 3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing: contained in the international application in written form. illed together with the international application in computer readable form. furnished subsequently to this Authority in written form. furnished subsequently to this Authority in computer readable form.

☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in

☐ The statement that the information recorded in computer readable form is identical to the written sequence

the international application as filed has been furnished.

 With regard to the elements of the international application (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed"

Form PCT/IPEA/409 (Boxes I-VIII, Sheet 1) (July 1998)

the description.

☐ the claims.

the drawings.

listing has been furnished.

4. The amendments have resulted in the cancellation of:

pages:

sheets:

Nos.:

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/CA00/01017

5. Mathia This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

see separate sheet

- 6. Additional observations, if necessary:
- V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- 1. Statement

Novelty (N)	Yes: No:	Claims Claims	1-25
Inventive step (IS)	Yes: No:	Claims Claims	1-25
Industrial applicability (IA)	Yes: No:	Claims	1-25

Citations and explanations see separate sheet

### VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made: see separate sheet

### **EXAMINATION REPORT - SEPARATE SHEET**

### Box I

- The amendments as filed with letter of 8.2.2001 are not allowable acc. Art. 19(2)
   PCT. <u>The present opinion has been drafted as if these amendments had not be n</u> made.
- 1.1 The amendment introducing a lower limit of 1 part per weight of elastomer in claims 1, 9 and 13 is not disclosed in the application as originally filed. The amendment to claim 13, "free of a tackifier" was not disclosed in the application as originally filed.
- 1.2 If the Applicant introduced the amendment as being a disclaimer, the following is noted: Claim 1 is not rendered novel over prior art by introducing an elastomer. Example 3 of D1 comprises Krayton G and ethylene/propylene copolymer which are both regarded as elastomers next to a functional polyolefin (maleic anhydride grafted polyethylene or EVA) and PS (a polystyrenic). The amendment to claim 1 if regarded as disclaimer hence is not establishing novelty and hence not allowable.
- 1.3 Claim 9 if amended by the introduction of elastomer would not be novel over D2, example 4 comprising 12 parts maleic anhydride grafted polyethylene, 30 parts EPR elastomer and 5 parts PS. Polymer PS is described on page 5, lines 6-23 and is regarded to include high impact polystyrene. The amended claim does also not appear to be novel over D5.
- 1.4 Claim 13 is not regarded as novel if it comprises elastomer and is free of tackifier. A composition comprising 1-30 parts of an elastomer, 5-30 parts high impact polystyrene and 5-95 parts functional polyolefin is known from D3: example A discloses compositions of 25% high impact polystyrene, 67% EVA (is regarded as a functional polyolefin) and 8% KRATON FG1901X (a maleated rubber, which is regarded as an elastomer. Hence even a combination of the amendments made is not establishing novelty of claim 13 over D3.
- 1.5 A tackifier is regarded as a polymer increasing tack. Functional polyolefin or grafted polyethylene hence could also be included in the definition for a tackifier. The tackifier in claim 13 hence would not be unambigously distinguishable from polymer b)

### Box V

- 1. The following documents are referred to in the present opinion:
  - D1 EP-A-412503
  - D2 EP-A-879862
  - D3 EP-A-712915
  - D4 EP-A-188901
  - D5 JP-A-61296044 (abstract)
  - D6 JP-A-61014272 (abstract)
  - D7 GB-A-2107325
  - D8 JP-A-59083651 (abstract)

The numbering will be adhered to in the rest of the procedure.

- 2. Claims 1-25 are regarded to not be novel in the sense of Art. 33(2) PCT
- 2.1 Subject matter of present claim 1 ist a composition of
  - 5-95 parts functional polyolefin
  - 5-40 parts polystyrenic resin

Such a composition is descibed in D1 (example 3), D2 (example 5), D3 (comparative example 1, page 4, line 12-14), D4 (example 11), D5 (abstract), D6 (abstract) and D7 (examples 8-12 and 15).

- 2.2 Subject matter of present claim 20 is an adhesive and subject matter of claim 23 a metal/adhesive/polymeric layer structure using the composition of claim 1. D1-D7 all describe adhesive compositions, in D1 (page 7, lines 21-27), D5, D6, D7 they are used for metal/resin laminates. Claims 20 and 23 are therefore not novel.
- 2.3 The subject matter of claims 2-8 dependant on claim 1 is not novel since the subject matter of
- a) claim 2 is disclosed in all of D1-D7
- b) claim 3 and 8 is disclosed in D2 (see page 7, line 29) and D3
- c) claim 4 is disclosed in D1 and D5
- d) claim 5 is disclosed in D1, D2, D5-D7
- e) claim 6 is disclosed in D1, D2, D5-D7
- f) claim 7 is disclosed in D1, D2, D4-D7

## **EXAMINATION REPORT - SEPARATE SHEET**

- g) claim 12 is disclosed in D1-D7
- 2.4 All other claims refer to (c)=impact modified polystyrene. Besides that the term is not allowable under Art. 6 PCT (see Box V), claims 9, 13-16 and 21 are also not novel:
- a) claim 9 and 21 are disclosed in D3 (comparative example 1, page 4, lines 12-14) and also D2 (page 5, line 10 and page 7, line 29)
- b) claim 13, 14, 15, 16 and 22 are disclosed in D2, example 5 and D3 (comparative example 1, page 4, lines 12-14). If EVA is regarded as a functional polyolefin, also the inventive examples, for example A, are novelty destroying to claim 13, since the KRATON FG1901X is an elastomer, even if it is modified.
- 2.5 The novelty of claims 10, 11, 17-19, 24 and 25 can only be decided upon, when there is a clear definition for high-impact polystyrene and a clear distinction between elastomer (d) and the other polymers of the composition
- 2.6 Moreover, the applicant did not proof that his claims are novel over D5. D6 and D8. which might contain novelty destroying subject matter for the claims mentioned under 2.5 in the full discosure.
- 3. However, claims 10, 11, 17-19, 24 and 25 do not appear to be inventive in respect to Art. 33(3) PCT
- 3.1 The above mentioned claims concern special embodiments of the invention relating to a special choice of a) (polyethylene, claim 10 and 11), c) (high-impact polystyrene, claims 10, 11, 17-19) and the relative amounts of the polymers (claim 17). There are no technical effects achieved by the addition of polyethylene or high-impact polystyrene as compared to EVA or other styrene polymers. The problem to be solved by the compositions claimed by claims 10, 11 and 17-19 can thus only be regarded as to provide alternative adhesive compositions bonding metal to polymers. However, similar such adhesive compositions comprising high-impact polystyrene (D2, D3) and polyethylene (D4 .D7) are known to somebody skilled in the art. At present, claims 10. 11, 17-19 hence have to be regarded as obvious for the skilled man since the applicant did not show, i.e by comparative examples, that high-impact polystyrene has a technical effect over other styrene polymers and polyethylene a technical effect over

## **EXAMINATION REPORT - SEPARATE SHEET**

EVA to define a technical problem

3.2 The subject matter of claims 24 and 25 are not inventive since the compositions. they are relating to, are not inventive, since it is obvious to somebody skilled in the art that all the compositions of D1-D7 will have at least some bond strength to metal or polymer films

### Box VIII

- 4.1 The term "high impact polystyrene" renders claims 3,9,10,13 and 17 unclear. Even if it would be chemically clear if high impact polystyrene can be regarded as a graft- or blockpolymer or as a fine physical blend of polystyrene with a rubbery polymer before blending, somebody skilled in the art will certainly not be able to distinguish between these possibilities in the claimed finally blended mixture. In the final adhesive blend, somebody skilled in the art will never be able to distinguish between a blend of polystyrene and EPR as used in D1 and an impact polystyrene of a similar composition added as such like in D2 or D3.
- 4.2 Claims 1, 10 and 13 are unclear, because the term "elastomer compound" does not distinguish compound (d) sufficiently from compound (a) and (c), which can be elastomeric as well
- 4.3 The numbering of the different compounds in claims 1, 10, 13 and 17 (a)-(f) is unclear. Some of the letters appear double or intermittant letters are missing (e)
- 4.4 The term "non-olefin" copolymer in claim 13 is linguistically unclear. It can also refer to other copolymers like polyamides, which are not supported by the description
- 4.5 The term "functional polyolefin" does not exclude non-grafted polyolefins with functional groups, i.e. EVA (claim 1, 13). Compound (b) hence is not distinguishable from compound (a), which can also be EVA. The ranges of (a) and (b) are hence also rendered unclear

### (19) World Intellectual Property Organization International Bureau



### (43) International Publication Date 15 March 2001 (15.03.2001)

### (10) International Publication Number WO 01/18141 A1

- (51) International Patent Classification?: C09J 123/08. [CA/CA]; 494 Grandtrunk Avenue West, Kingston, Ontario K7M 8P6 (CA). TANNY, Stephen, Robert [US/US]; 111 123/16, B32B 27/08 Horseshoc Road, Newark, DE 19711-2412 (US). (21) International Application Number: PCT/CA00/01017 (74) Agents: CLARIZIO, Dino, P. et al.; Dimock Stratton Clarizio LLP, 20 Queen St. W., Suite 3202, Box 102, (22) International Filing Date: Toronto, Ontario M5H 3R3 (CA). 1 September 2000 (01.09.2000) English (81) Designated States (national): CA, CN, JP, KR, US. (25) Filing Language: (26) Publication Language: English (84) Designated States (regional): European patent (AT, BE, CH. CY. DE, DK. ES. FI, FR. GB, GR. IE, IT, LU, MC. ·NL, PT, SE). (30) Priority Data: 60/152,539 3 September 1999 (03.09.1999) US
- (71) Applicant (for all designated States except US): E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 - With amended claims. Market Street, Wilmington, DE 19898 (US).

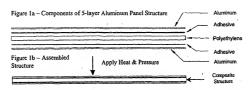
With international search report.

Published:

(72) Inventors; and (75) Inventors/Applicants (for US only): HOJABR, Sassan ning of each regular issue of the PCT Gazette.

For two-letter codes and other abbreviations, refer to the "Guidance Notes an Cades and Abbreviations" appearing at the begin-

(54) Title: LOW ACTIVATION TEMPERATURE ADHESIVE COMPOSITION WITH HIGH PEEL STRENGTH AND COHE-SIVE FAILURE



(57) Abstract: The present invention provides a resin composition made from admixing starting materials comprising: (a) 0 to 90 parts by weight of a polyolefin; (b) 5 to 95 parts by weight of a functional polyolefin; (c) 5 to 40 parts by weight of a polystyrenic; and (d) 0 to 30 parts by weight of an elastomer, where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight. In a further embodiment, component (a) is a non-olefin copolymer and component (c) is high impact polystyrene. The resin compositions are useful as adhesives for metal and polyolefin substrates.

WO 01/18141 PCT/CA00/01017

1

# Low Activation Temperature Adhesive Composition With High Peel Strength and Cohesive Failure

### Field of the Invention:

This invention relates to new resin and adhesive compositions, and in particular to

low activation temperature adhesive compositions that can be applied as a selfsupporting film or can be co-extruded or extrusion coated onto a substrate.

### Background of the Invention:

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While it is known that acid-modified or anhydride-modified polymers are useful as adhesives for bonding to both metal and polyolefins, there is a need to have adhesives with not only high-bond strength to both metal and polyolefins, but also to activate the bonding at relatively lower temperatures. Conventional and currently commercially available modified polyolefins sacrifice either bonding strength to the metal or to the polyolefin, or do not activate at low temperatures when bonding to metals or polyolefins. Furthermore, adhesive compositions that purport to activate at low temperatures are often very soft and tacky materials such that the handling of these compositions as free films in thermal laminating processes is problematic and may require very costly release films to prevent blocking. Also, most current adhesives tend to significantly lose their bond strength over time after they are put into service. This phenomenon is referred to as "age-down" in the industry.

There are number of patents in the art that disclose adhesive compositions but these prior art patents are not satisfactory in every aspect mentioned in this specification. Examples of these prior art patents include United States Patent Nos. 5,516,583; 4,861,676; 4,861,677; 4,552,819; and 5,965,255.

One example of the prior art is United States Patent No. 5,225,482 issued July 6, 1993
25 to Nakagawa et al, which discloses an adhesive composition comprising an ethylenevinyl acetate copolymer, a styrene polymer resin, a graft-modified polyethylene, a
polystyrene elastomer and an ethyleneα-olefin coploymer. There is no disclosure of
the use of high impact polystyrene as the styrene resin in combination with an
olefin/non-olefin copolymer.

It has been found that the formulations for the adhesive compositions of the present invention solve many, or all, of these problems. The adhesive compositions of the present invention bond to both metal and polyolefins, activate at relatively lower temperatures and are easily handled as a free or coextruded film not requiring

5 interleafing or a release paper. In addition, it has been found that these adhesive compositions fail 100% cohesively during the peel test. Cohesive failure is a desirable attribute in that it can be an indicator of high bond strength, so much so that the bond strength is greater than the cohesive strength of the adhesive. Cohesive failure also provides a convenient visual test to assure that multi-layer constructions are properly bonded when other testing methods are not readily available. Finally there appears to be a strong correlation between the cohesive failure mode of the adhesive and the retention of bond strength after the product is put into service.

Accordingly, the present invention provides adhesive compositions that possess excellent bonding properties to metallic substrates and to a number of polymeric materials resulting in an adhesive layer with high peel strength. The present invention also allows for a relatively lower activation temperature to be used in the manufacture of building panels.

The adhesive compositions of the present invention may be supplied as an adhesive resin in pellet form or as an adhesive film.

### 20 Summary of the Invention:

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Accordingly, in one aspect of the present invention, there is provided a resin composition made from admixing starting materials comprising:

- (a) 0 to 90 parts by weight of a polyolefin;
- (b) 5 to 95 parts by weight of a functional polyolefin;
- (c) 5 to 40 parts by weight of a polystyrenic; and
- (d) 0 to 30 parts by weight of an elastomer.

In a second aspect of the present invention, there is provided a resin composition made from admixing starting materials comprising:

- (a) 0 to 90 parts by weight of a non-olefin copolymer:
- (b) 5 to 95 parts by weight of a functional polyolefin;
- (c) 5 to 40 parts by weight of high impact polystyrene; and
  - (e) 0 to 30 parts by weight of an elastomer,

where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.

### Brief Description of the Drawings:

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10 The preferred embodiments of the present invention will be described with reference to the accompanying drawings in which like numerals refer to the same parts in the several views and in which:

Figures 1a and 1b are schematic views of a five-layer laminated structure made using one embodiment of the adhesive composition of the present invention.

### 15 Detailed Description of the Preferred Embodiments:

The present invention will be described with reference to its preferred embodiments.

The preferred resin and adhesive compositions of the present invention as described herein are melt blends of a number of polymers. These adhesives can be used in various kinds of applications and possess varying properties depending on the specific application. One application for these preferred adhesives is in manufacturing metal panels for the construction industry in which the panels are in the form of a composite structure comprising metal substrates such as aluminum or steel bonded to a polyolefin core such as polyethylene. The preferred adhesive compositions of the present invention are used to bond the metal substrates to the polyethylene core.

25 In such an application, it is important for the adhesive layer to provide excellent adhesion of the polyolefin to the metal substrate. As well, it is desirable that the adhesive fail cohesively during application, and should be relatively easy to formulate and use. It has been found that the adhesive compositions of the present invention

WO 01/18141 PCT/CA00/01017

4

provide excellent adhesion to metallic substrates and to a number of polymeric materials. They fail 100% cohesively during the peel test. The adhesive compositions can be used as self-supporting films, making them easy to handle and process.

The preferred adhesive compositions of the present invention activate at lower temperatures than adhesives currently used in manufacturing building panels. For the preferred adhesive compositions, the activation temperature can be lowered down to about 125 °C. This lowering of the activation temperature results in considerable cost savings and safer operations for panel manufacturers because the use of protective layers for painted surfaces (which are needed at higher temperatures to prevent mottling of the paint) can be eliminated from the manufacturing process.

In one aspect, the resin composition of the present invention comprises:

a) 0 to 90 parts by weight of a polyolefin;

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- b) 5 to 95 parts by weight of a functional polyolefin;
- 5 to 40 parts by weight of a polystyrenic; and
- d) 0 to 30 parts by weight of an elastomer.

where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.

In a second aspect, the resin composition of the present invention comprises:

- a) 0 to 90 parts by weight of a non-olefin copolymer;
- b) 5 to 95 parts by weight of a functional polyolefin;
- c) 5 to 40 parts by weight of high impact polystyrene; and
- e) 0 to 30 parts by weight of an elastomer,

where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.

25 In this patent specification, the term "polyolefin" means homopolymers and copolymers of olefins. More specifically, homopolymers include polymers consisting of a single unsaturated olefin such as polyethylene, polypropylene, polybutene or the like where the olefin has 2-20 carbon atoms. Copolymers of olefins include polymers

consisting of one or more unsaturated or multiply unsaturated hydrocarbons having 2-20 carbon atoms. Examples include, but are not limited to ethylene/propylene copolymers, ethylene/butene copolymers, ethylene/butene copolymers, ethylene/octene copolymers, ethylene/styrene copolymers, ethylene/butene/octene copolymers, ethylene/propylene/norbornadiene copolymers and propylene/butene copolymers.

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In this patent specification, the term "non-olefin copolymer" refers to copolymers of olefins with non-olefins. Non-olefins that can be copolymerized with olefins, principally ethylene, include but are not limited to: vinyl acetate, acrylate or methacrylate esters having 1-20 carbon atoms, unsaturated anhydrides such as maleic or itaconic anhydride, unsaturated acids such as maleic, fumaric, acrylic, methacrylic or itaconic acid. Examples of copolymers of olefins and non-olefins include, but are not limited to: ethylene/vinyl acetate, ethylene/methylacrylate, ethylene/butylacrylate. These polymers can be made by processes well known in the art, including the use of metallocene catalysts, Ziegler Natta catalysts and other catalysts useful in "low pressure" polymerization processes. Conversely, these polymers may be made in "high pressure" polymerization processes using, for example, free radical initiators. Mixtures and blends of the Polyolefins may be used.

In this patent specification, the term "functional polyolefin" refers to a polyolefin or a

20 non-olefin copolymer that has specific functional groups capable of reacting to form
covalent or ionic bonds. A functional polyolefin includes a "grafted polyolefin" as
defined below.

The term "grafted polyolefin" refers to a polyolefin, to a non-olefin copolymer or to a mixture or blend of polyolefins and/or non-olefin copolymers, onto which is grafted at least one monomer selected from ethylenically unsaturated carboxylic acids and ethylenically unsaturated carboxylic acid anhydrides, including less preferably, derivatives of such acids, and mixtures thereof. Examples of the acids and anhydrides, which may be mono-, di- or polycarboxylic acids are acrylic acid, methacrylic acid, maleic acid, fumaric acid, itaconic acid, crotonic acid, itaconic anhydride, maleic anhydride and substituted maleic anhydride, e.g. dimethyl maleic anhydride or

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citrotonic anhydride, nadic anhydride, nadic methyl anhydride, and tetrahydrophthalic anhydride, maleic anhydride being particularly preferred. Examples of the derivatives of the unsaturated acids are salts, amides, imides and esters, e.g., mono- and disodium maleate, acrylamide, glycidyl methacrylate and dimethyl fumarate. Grafted polyolefins are well known in the art and can be produced by a variety of processes including thermal grafting in an extruder or other mixing device, grafting in solution or grafting in a fluidized bed reactor. Blends or mixtures of grafted polyolefins may also be used.

In this patent specification, the term "polystyrenic" refers to homopolymers of

stryrene or alpha methylstyrene or copolymers of styene with unsaturated monomers

such as, but not limited to: ethylene, butene, butadiene, or isoprene. Specific examples
include, but are not limited to: ethylene/styrene random or block copolymers,
ethylene/butadiene random or block copolymers and hydrogenated and partially
hydrogenated butadiene/styrene copolymers. Also useful are polystyrenics further

modified for enhanced impact properties and usually referred to as High Impact
Polystyrene or HIPS. Blends and mixtures of polystyrenics may also be used.

Specific examples include, but are not limited to, High Performance Stryrenics sold by
Nova Chemicals and ethylene/styrene copolymers sold by Dow Chemical under the
name Index<sup>TM</sup>.

In this patent specification, the term "elastomer" also refers to a polyolefin or polystyrenic but is dinstinguised from polyolefin or polystyrenic as described above in having a relatively low level of crystallinity, that is, in being relatively amorphous. Elastomers by definition herein will have a heat a fusion, as measured by Differential Scanning Calorimetry (DSC), at 10 degrees centigrade per minute heating rate, of less than 30 joules/gram. A polyolefin elastomer, for example, may be a copolymer of ethylene and an alpha olefin and would include low density metallocene ethylene/butene copolymers sold by ExxonMobil under the name "Exact®" or metallocene ethylene/octene copolymers sold by DuPont Dow Elastomers under the name "Engage®". It would also include ethylene/propylene copolymers sold by ExxonMobil under the name "Vistalon®", ethylene/alpha olefin copolymer sold by

WO 01/18141 PCT/CA00/01017

7

Mitsui under the name "Tafiner®" or ethylene/propylene/norbornadiene copolymers sold by DuPont Dow Elastomers under the name of "Nordel®". It would also include polybutene rubber, polyisobutylene. Examples of a polystyrenic elastomer would include, for example, di-block and tri-block copolymers sold by Shell under the name "Kraton®" or those sold by Firestone under the name "Stereon®".

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In addition to the above-mentioned components, the preferred adhesive compositions of the present invention may contain small amounts of other materials commonly used and well known in the adhesive art. These materials include, for example, primary and secondary antioxidants, stabilizers, slip additives, antiblock additives such as silica or tale, dyes, pigments and tackifying resins such as those described in the Kirk Othmer Encyclopedia of Chemical Technology, so long as the addition of these additives does not significantly adversely affect the adhesive qualities of the composition.

The adhesive compositions of the present invention can be dry blended and subsequently melt blended in a twin screw extruder and repelletized as is well known in the art. Subsequently, these melt blended resins can be converted and applied by a variety of techniques and processes. For example, the adhesive can be converted into a film by cast or blown film die extrusion techniques and this adhesive film can be laminated to appropriate substrates such as metals or polyolefins. As an alternative, the adhesive composition can be coextruded with other polyolefins as a skin layer on either one or both surfaces of the polyolefin to produce a more economical adhesive film.

As a further alternative, a coextruded film can be created incorporating polar barrier resins such as polyamides, ethylene vinyl alcohol copolymer (EVOH) or polyester using the adhesive compositions of the present invention to bond directly to the polar barrier material. These adhesive films can be laminated to various substrates by heat activating the adhesive film. Heat activation can be done by a variety of methods including, but not limited to, direct contact with a heated plate or roller, absorption of infrared energy, direct heating in an oven or activation through RF frequency or microwave radiation.

WO 01/18141 PCT/CA00/01017

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In another application for the adhesive compositions of the present invention, the adhesive can be directly coated onto a substrate in processes well known in the art, including, for example, extrusion lamination, extrusion coating, coextrusion lamination and coextrusion coating. The adhesive composition of the present invention can be used to bond to polar barrier resins, such as EVOH, polyamide or polyester. It can also be used to bond to metals, such as steel, aluminum, copper and brass, and to polyolefins such as polyethylene, ethylene copolymers and polypropylene.

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In one preferred embodiment of the present invention, a resin composition is provided
made from starting materials that comprise the following components:

- (a) 0 to 90 parts by weight, and more preferably 20 to 60 parts by weight, of linear low density polyethylene (for example, commercially available under the name Exact® from ExxonMobil or under the name Sclair® from Nova Chemicals);
- (b) 5 to 95 parts by weight, and more preferably 10 to 30 parts by weight, of maleic anhydride grafted polyethylene (for example, commercially available under the name Fusabond® from DuPont);
  - (c) 5 to 40 parts by weight, and more preferably 10 to 35 parts by weight, of high impact polystyrene (for example, commercially available as High Performance Styrenics from Nova Chemicals); and
  - (d) 0 to 30 parts by weight, and more preferably 10 to 25 parts by weight, of an ethylene-propylene diene rubber compound (for example, commercially available under the name Nordel IP® from DuPont-Dow Elastomers);
  - where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.

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In a second embodiment of the present invention, an adhesive composition is provided that comprises the following components:

- (a) 0 to 90 parts by weight, and more preferably 40 to 60 parts by weight, of ethylene vinyl acetate copolymer preferably having 3 to 40 wt% of vinyl acetate, and more preferably, 5 to 30 wt% of vinyl acetate (an example of a suitable commercially available ethylene vinyl acetate copolymer is sold under the name Elvax® by DuPont);
- (b) 5 to 95 parts by weight, and more preferably 10 to 30 parts by weight, of maleic anhydride grafted polyethylene (for example, commercially available under the name Fusabond ® from DuPont);
- (c) 5 to 40 parts by weight, and more preferably 10 to 35 parts by weight, of high impact polystyrene (for example, commercially available as High Performance Styrenics from Nova Chemicals); and
- (d) 0 to 30 parts by weight, and more preferably 10 to 25 parts by weight, of an ethylene-propylene diene rubber compound (for example, commercially available under the name Nordel IP® from DuPont-Dow Elastomers);

- 20 In these two preferred embodiments of the present invention, polyisobutylene may be used instead of the ethylene-propylene diene elastomer compound. As well, polystyrenic styrene-butadiene elastomer having different butadiene contents may be used with similar effects.
- Each component of the adhesive compositions of the present invention preferably

  25 provides certain attributes to the final adhesive. The low melting point polyolefin,
  such as polyethylene or ethylene vinyl acetate copolymer, enhances the low activation
  temperature property of the adhesive. The grafted polymer mainly contributes to the
  superior bonding of the adhesive composition to metal substrates. The elastomer
  component enhances its resistance to debonding and increases toughness of the final

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adhesive compositions. Finally, the polystyrenic component provides the superior cohesive failure properties, as well as, surprisingly, higher bonding properties.

The preferred adhesive compositions of the present invention can have melt flow rates between 0.1 to 100 dg/min, more preferably from 0.5 to 50 dg/min and most preferably from 0.8 to 25 dg/min., as measured by ASTM-1238E. The peak melting point, as measured by DSC, can be between 55 °C- 140 °C depending on the composition.

Because of their unique combination of high peel strength, low surface tack when processed into films and bonding at low temperature, the preferred adhesives of the present invention are useful in the manufacture of aluminum and steel building panels.

### Examples:

The following examples show the surprising advantages of using a polystyrenic such as high impact polystyrene (HIPS) in providing additional peel strength, cohesive mode of failure and a non-tacky adhesive film.

### Example 1:

Table 1 shows the compositions 1A, 1B and 1C at 0 wt%, 10 wt% and 20 wt% High Impact Polystyrene ("HIPS") respectively. These compositions were dry-blended initially and fed to a small co-rotating twin-screw extruder. The melt-compounded materials were pelletized and then blown into film with an average thickness of about 75 microns. The films were subsequently used as adhesive layers to make a 5 layer composite structure as shown in Figures 1a and 1b. The aluminium was aludyne treated and had a thickness of 0.38 mm. The polyethylene core used was low-density polyethylene (LDPE) sheet, 2 mm thick.

- 25 The lamination of the structures were carried out in a electrically heated press as follows (Figure 1a and 1b):
  - 1- Preheat the 5-layer structure for 4.5 min. @ 100 °C.
  - 2- Apply pressure of 5 kgf/cm<sup>2</sup> at 135 °C for 20 seconds.

WO 01/18141 PCT/CA00/01017

11

- 3- Release the pressure and leave the composite in the press for an additional
- 1.5 min. @ 135°C.
- 4- Air cool the sample to room temperature.

The composite structure was then tested to measure the peel strength (ASTM 1876)

5 using an Instron™ machine. The following conditions were used during the peel strength test:

- i) Crosshead speed: 100 mm/min.
- ii) Peel mode: 180º

The last two columns in Table 1 indicate the results of the peel strength and failure

mode as the amount of Component d (HIPS) is increased from 0 wt% to 20 wt%.

Comparing composition 1A with compositions 1B and 1C, the surprising effect of peel strength enhancement due to the presence of the polystyrenic HIPS component can be seen. It is also highly desirable to achieve a cohesive failure (50/50 to polyethylene and aluminum respectively). Sample 1C shows 80% cohesive failure as

compared to adhesive failure in the other two compositions.

Table 1: MonoLayer Ethylene Vinyl Acetate (EVA) based Adhesives

	_	_	_
Palline Mode	Adhesive to Aluminum	Adhesive to Aluminum	80% Cohesive
Peel Strength Keft Smith	16.4	23.6	29.8
Gomp. (8) Wes	0.1	0.1	0.1
Comp.(I) Wrog.	0.0%	10.0	20.0
Comp. (c) W.C.o.	20.0	20.0	20.0
Comp.(b)	30.0	30.0	30.0
Comp.(a) W192	49.9	39.9	29.9
D.Ne.	14	118	21

Component (b) = Linear low-density polyethylene (LLDPE) grafted with maleic anhydride at 0.9wr% and MI of 2.5 dg/min. Component (c) = Ethylene/propylene/norbornadiene copolymers (EPDM) Elastomers with Moony Viscosity of 20. Component (a) = Ethylene vinyl acetate (EVA) copolymer with 12% VA content and MI of 2.5 dg/min. Component (d) = Polybutadiene impact modified polystyrene (HIPS) Elastomers

Component (e) = Hindered polyphenol antioxidant stabilizer

### Example 2:

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Samples were prepared as described in Example 1 with the exception that co-extruded blown films of polyethylene/Adhesive were made and substituted for a pure adhesive film in order to reduce the cost of the adhesive used. The compositions 2A, 2B and 2C are co-extruded with a linear low density polyethylene (LLDPE) producing LLDPE-Adhesive (each layer being 25 micron thick). The results are summarized in Table 2. The co-extruded films generally result in lower peel strength as can be seen comparing 1C vs. 2A (29.8 vs. 16.5 kgf/25mm, respectively) but they are still functional. The addition of high impact polystyrene at 30wt% level (2C) appears to lower the peel strength. This latter composition is still functional though not optimal.

Table 2: 2-Layer Ethylene Vinyl Acetate (EVA)/ Polyethylene based Adhesives

ğ	100% Cohesive	100% Cohesive	100% Cohesive
Ž.	ohes	ohes	ohes
	%	%	%
H	음	음	9
		-	
3.3	- 1		
2.2	16.5	17.5	10.9
		-	_
e .			
			_
	0.1	9	0
		T	
	- 1	ł	
8.5	20.0	20.0	30.0
	7	7	~
2		- 1	
22			0
3	20.0	15.0	5.
	+	-	
		ı	
题	30.0	20.0	20.0
	ñ	7	7
3.			
	6	6	6
Ğ.	29.9	44.9	34.9
127400	-	-	
2	χ	2B	20
	7	2	7

Component (b) = High-density polyethylene (HDPE) grafted with maleic anhydride at 1.0 wt% and MI of 11 dg/min. Component (c) = Ethylene/propylene/norbornadiene copolymers (EPDM) Elastomers with Moony Viscosity of 20. Component (a) = Metallocene ethylene butene copolymer with 0.905 density and MI of 4.5 dg/min. Component (d) = Polybutadiene impact modified polystyrene (HIPS) Elastomers Component (e) = Hindered polyphenol antioxidant stabilizer

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### Example 3:

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In this example, compositions were made using metallocene linear low-density polyethylene as the base resin. Table 3 summarises the compositions. Samples 3A, 3B and 3C were prepared as those described in Examples 1 and 2. The last two compositions 3D and 3E were prepared in a 2-step process, in order to more closely simulate an actual commercial production. Adhesives of 3D and 3E were initially laminated to aluminium at a temperature of 149 °C, using a pressure of 8.5 kgf/cm<sup>2</sup> for 30 seconds. These pre-laminated aluminium sheets were then pressed with a pre-heated LDPE core at 132 °C, using a pressure of 8.5 kgf/cm<sup>2</sup> for 10 seconds. The peel testing was performed at a 90° angle as opposed to the 180° angle used in the other examples. Composition 3D is a repeat of 3C and is used to compare the performance between the different lamination and testing methods. Sample 3D represents much lower peel strength than 3C due to 180° peel test vs. a 90° peel for sample 3C.

15 Once again increasing the amount of impact modified polystyrene (HIPS) from 0wt% - 20wt% (comparing samples 3A to 3C) resulted in significant increase in peel strength. The mode of failure also improved and become more cohesive. Sample 3E, containing 30% HIPS, shows a high peel strength value (20.5 kgf/25 mm in 90° peel mode) and 00% cohesive failure. It should be noted that products currently used in 20 commercial production totally fail when using this "low lamination temperature" of about 132°C.

Table 3: Metallocene LLDPE based Adbesives:

I venture i ve				_	
Failure Mode	Adhesive To Aluminum	Adhesive To Aluminum	20% Cohesive	30% Cohesive	100% Cohesive
P. Kelisteneth	N/A	26.9	36.3	18.6	20.5
(6) (0) (0) (0) (0) (1) (1) (1) (1) (1) (1) (1) (1) (1) (1	0.10	0.10	0.10	0.10	0.10
To Manager	0.0	10.0	20.0	20.0	30.0
Solution (C)	20.0	20.0	20.0	20.0	20.0
Count ()	20.0	20.0	20.0	20.0	20.0
Comp. (4)	59.9	49.9	39.9	39.9	29.9
ID No	3A	38	၁င	30	3E

Component (b) = High-density polyethylene (HDPE) grafted with maleic anhydride at 1.0 wt% and MI of 11 dg/min. Component (c) = Ethylene/propylene/norbornadiene copolymers (EPDM) Elastomers with Moony Viscosity of 20. Component (a) = Metallocene ethylene butene copolymer with 0.905 density and MI of 4.5 dg/min.

Component (d) = Polybutadiene impact modified polystyrene (HIPS) Elastomers Component (e) = Hindered polyphenol antioxidant stabilizer

2

WO 01/18141 PCT/CA00/01017

17

Although the present invention has been shown and described with respect to its preferred embodiments, it will be understood by those skilled in the art that other changes, modifications, additions and omissions may be made without departing from the substance and the scope of the present invention as defined by the attached claims.

### What is Claimed is:

- 1. A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of a polyolefin;
  - (b) 5 to 95 parts by weight of a functional polyolefin;
  - (c) 5 to 40 parts by weight of a polystyrenic; and
  - (f) 0 to 30 parts by weight of an elastomer,

- The resin of claim 1 wherein the polystyrenic is selected from the group consisting of a homopolymer of styrene and a copolymer of styrene with unsaturated monomers.
- The resin of claim 1 wherein the polystyrenic is high impact polystyrene.
- The resin of claim 1 wherein the polysterenic is a blend of two or more different polystyrenics.
- The resin of claim 1 wherein the polystyrenic is a copolymer of styrene with a
  monomer selected from the group consisting of ethylene, butene, butadiene or
  isoprene.
- 6. The resin of claim 5 wherein the polystyerenic is selected from the group consisting of ethylene/styrene random or block copolymers, ethylene/butadiene random or block copolymer and and hydrogentaed and partially hydrogeated butadiene/styrene copolymers.
- The resin composition of claim 1, wherein the functional polyolefin is a grafted polyolefin.
- The resin composition of claim 3, wherein the functional polyolefin is a grafted polyolefin.

- 9. A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of polyethylene;
  - (b) 5 to 95 parts by weight of maleic anhydride grafted polyethylene;
  - (c) 5 to 40 parts by weight of high impact polystyrene; and
  - (d) 0 to 30 parts by weight of an elastomer selected from the group consisting of ethylene-propylene elastomer, ethylene-propylene diene elastomer, polyisobutylene and polyisobutylene styrene-butadiene elastomer,

where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.

- 10. The resin composition of claim 9 made from admixing starting materials comprising:
  - (a) 40 to 60 parts by weight of polyethylene;
  - (b) 10 to 30 parts by weight of maleic anhydride grafted polyethylene;
  - (c) 10 to 35 part s be weight of the high impact polystyrene; and
  - (c) 10 to 25 parts by weight of the elastomer compound,

- The resin composition of claim 10 wherein the polyethylene comprises one or more of high density polyethylene, linear low density polyethylene and low density polyethylene.
- 12. The resin composition of claim 7 wherein the grafted polyethylene is selected from the group consisting of maleic anhydride grafted linear low density polyethylene, maleic anhydride low density polyethylene, maleic anhydride grafted high density polyethylene.
- 13. A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of a non-olefin copolymer;

- (b) 5 to 95 parts by weight of a functional polyolefin;
- (c) 5 to 40 parts by weight of high impact polystyrene; and
- (g) 0 to 30 parts by weight of an elastomer,
  where the total amount of components of (a), (b), (c) and (d) in the resin
  composition is 100 parts by weight.
- 14. The resin composition of claim 13, wherein the non-olefin copolymer is ethyelene vinyl acetate copolymer.
- 15. The resin composition of claim 13 wherein the grafted polyethylene is selected from the group consisting of maleic anhydride grafted linear low density polyethylene, maleic anhydride grafted low density polyethylene and maleic anhydride grafted high density polyethylene.
- The resin composition of claim 15, wherein the functional polyolefin is maleic anhydride grafted polyethylene.
- 17. The resin composition of claim 13, comprising:
  - (a) 40 to 60 parts by weight of ethylene vinyl acetate copolymer;
  - (b) 10 to 30 parts by weight of maleic anhydride grafted polyethylene;
  - (d) 10 to 35 parts by weight of high impact polystyrene; and
  - (d) 10 to 20 parts by weight of an elastomer compound. where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.
- The resin composition of claim 17 wherein the ethylene vinyl acetate copolymer contains 3 to 40wt% vinyl acetate.
- The resin composition of claim 18 wherein the ethylene vinyl acetate copolymer contains 5 to 30 wt% vinyl acetate.
- 20. An adhesive composition comprising the resin of claim 1.
- 21. An adhesive composition comprising the resin of claim 9.

- 22. An adhesive composition comprising the resin of claim 13.
- 23. A composite structure comprising:
  - (a) a metal substrate;
  - (b) a polymeric layer; and
  - (c) a layer of the adhesive composition of claim 20 between the metal substrate and the polymeric layer.
- 24. A composite structure comprising:
  - (a) a metal substrate;
  - (b) a polymeric layer; and
  - (c) a layer of the adhesive composition of claim 21 between the metal substrate and the polymeric layer.
- 25. A composite structure comprising:
  - (a) a metal substrate;
  - (b) a polymeric layer; and
  - (c) a layer of the adhesive composition of claim 22 between the metal substrate and the polymeric layer.

### AMENDED CLAIMS

[received by the International Bureau on 08 February 2001 (08.02.01) original claims 1, 9, 13 amended; remaining claims unchanged (3 pages)]

- A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of a polyolefin;
  - (b) 5 to 95 parts by weight of a functional polyolefin;
  - (c) 5 to 40 parts by weight of a polystyrenic; and
  - (d) 1 to 30 parts by weight of an elastomer,

- The resin of claim 1 wherein the polystyrenic is selected from the group consisting of a homopolymer of styrene and a copolymer of styrene with unsaturated monomers.
- 3. The resin of claim 1 wherein the polystyrenic is high impact polystyrene.
- The resin of claim 1 wherein the polysterenic is a blend of two or more different polystyrenics.
- The resin of claim 1 wherein the polystyrenic is a copolymer of styrene with a
  monomer selected from the group consisting of ethylene, butene, butadiene or
  isoprene.
- 6. The resin of claim 5 wherein the polystyerenic is selected from the group consisting of ethylene/styrene random or block copolymers, ethylene/butadiene random or block copolymer and and hydrogentaed and partially hydrogeated butadiene/styrene copolymers.
- The resin composition of claim 1, wherein the functional polyolefin is a grafted polyolefin.
- The resin composition of claim 3, wherein the functional polyolefin is a grafted polyolefin.

- 9. A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of polyethylene;
  - (b) 5 to 95 parts by weight of maleic anhydride grafted polyethylene;
  - (c) 5 to 40 parts by weight of high impact polystyrene; and
  - (d) 1 to 30 parts by weight of an elastomer selected from the group consisting of ethylene-propylene elastomer, ethylene-propyelene diene elastomer, polyisobutylene and polyisobutylene styrene-butadiene elastomer.

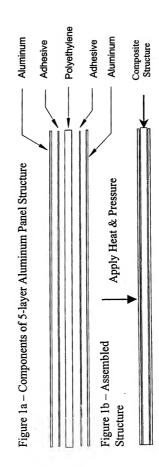
- 10. The resin composition of claim 9 made from admixing starting materials comprising:
  - (a) 40 to 60 parts by weight of polyethylene;
  - (b) 10 to 30 parts by weight of maleic anhydride grafted polyethylene;
  - (c) 10 to 35 part s be weight of the high impact polystyrene; and
  - (c) 10 to 25 parts by weight of the elastomer compound, where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.
- The resin composition of claim 10 wherein the polyethylene comprises one or more of high density polyethylene, linear low density polyethylene and low density polyethylene.
- 12. The resin composition of claim 7 wherein the grafted polyethylene is selected from the group consisting of maleic anhydride grafted linear low density polyethylene, maleic anhydride low density polyethylene, maleic anhydride grafted high density polyethylene.
- 13. A resin composition made from admixing starting materials comprising:
  - (a) 0 to 90 parts by weight of a non-olefin copolymer;

- (b) 5 to 95 parts by weight of a functional polyolefin;
- (c) 5 to 40 parts by weight of high impact polystyrene; and
- (e) 1 to 30 parts by weight of an elastomer,

where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight and wherein the resin composition is free of a tackifier.

- 14. The resin composition of claim 13, wherein the non-olefin copolymer is ethyclene vinyl acetate copolymer.
- 15. The resin composition of claim 13 wherein the grafted polyethylene is selected from the group consisting of maleic anhydride grafted linear low density polyethylene, maleic anhydride grafted low density polyethylene and maleic anhydride grafted high density polyethylene.
- The resin composition of claim 15, wherein the functional polyolefin is maleic anhydride grafted polyethylene.
- 17. The resin composition of claim 13, comprising:
  - (a) 40 to 60 parts by weight of ethylene vinyl acetate copolymer;
  - (b) 10 to 30 parts by weight of maleic anhydride grafted polyethylene;
  - (d) 10 to 35 parts by weight of high impact polystyrene; and
  - (d) 10 to 20 parts by weight of an elastomer compound. where the total amount of components of (a), (b), (c) and (d) in the resin composition is 100 parts by weight.
- The resin composition of claim 17 wherein the ethylene vinyl acetate copolymer contains 3 to 40wt% vinyl acetate.
- The resin composition of claim 18 wherein the ethylene vinyl acetate copolymer contains 5 to 30 wt% vinyl acetate.
- An adhesive composition comprising the resin of claim 1.

Figure 1:



## INTERNATIONAL SEARCH REPORT

Intern nal Application No PCT/CA 00/01017

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C09J123/08 C09J123/16 B32B27/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  $IPC\ 7\ C09J\ B32B\ C08L$ 

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

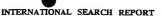
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Date of the actual completion of the international search  17 November 2000	Date of mailing of the international search report  08/12/2000
Name and mailing address of the ISA  European Patent Office, P.B. 5818 Patentiaan 2 N.L 2206 HY Rijsvijk Tel. (-31-70) 340-2040, Tx. 31 651 epo nl, Fax: (-31-70) 340-3016	Authorized officer Schmidt, H

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT Category \* | Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. χ DATABASE WPI 1-20 Derwent Publications Ltd., London, GB; AN 1987-040157 s XP002153231 & JP 61 296044 A (MITSUI), 26 December 1986 (1986-12-26) abstract X DATABASE WPI 1-20 Derwent Publications Ltd., London, GB: AN 1986-064319 ' XP002153232 & JP 61 014272 A (UBE). 22 January 1986 (1986-01-22) abstract EP 0 188 901 A (MITSUI PETROCHEMICAL IND) X 1-20 30 July 1986 (1986-07-30) examples 9-13 GB 2 107 325 A (ACC CHEM CO:GETTY CHEMICAL 1-20 CO) 27 April 1983 (1983-04-27) examples 8-12.15 X DATABASE WPI 1-20 Derwent Publications Ltd., London, GB; AN 1984-156105 XP002153233 & JP 59 083651 A (SUMITOMO). 15 May 1984 (1984-05-15) abstract

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